Laboratory of Polymer Physical Chemistry (Special Issue on the Commemoration of the Fiftieth Anniversary)

Bull. Inst. Chem. Res., Kyoto Univ., Vol. 54, No. 6, 1976

LABORATORY OF POLYMER PHYSICAL CHEMISTRY

Head: Dr. Michio Kurata

A list of scientific papers published from this laboratory was given in 1966 in the Commemoration Volume for the Fortieth Anniversary of the Institute for Chemical Research. The following list covers the papers published in the subsequent period 1966–1976.

The first nine papers (1–9) treat of theoretical problems in the field of polymer chain statistics and dynamics. Kurata has developed a new theory of the excluded volume effect of linear polymer chains which corresponds to the Bethe approximation of the lattice statistics, and presented an equation of a hybrid form between Flory's and Fixman's equations. This equation was derived independently by Alexandrowicz also and it has gained support from the recent Monte Carlo data. Iwata and Kurata presented a theory of polymer chain dynamics based on the irreversible statistical mechanics and were able to clarify the relation between the classical spring-beads model of Rouse and the local-jump model of Stockmayer and Verdier. This theory was then extended by Iwata to the analysis of chain motions in the high frequency region. Osaki has found a revised solution of the Zimm integrodifferential equation for polymer chain dynamics, and presented a numerical table which is useful for analysis of the viscoelastic properties of dilute solutions of linear and branched polymers.

Odani studied the sorption and desorption kinetics of some organic vapors by polymer films and presented an ingenious method for eliminating the film thickness effect in determination of true diffusion coefficient (10). His group has continued the study of permeation, diffusion, and solution of inert gases in block copolymer films in hope of elucidating the mechanism of permselectivity (11–13).

The viscoelastic properties of polymer systems has been major subjects of our study in this decade as will be seen from an abundance of papers in the following list. Based on the normal stress measurements in dilute polymer solutions, we were able to verify that polymer chains behave like non-draining coils at infinite dilution (14–16). This had been assumed in the treatment of intrinsic viscosity for many years without direct experimental evidence. The viscoelastic properties of dilute polymer solutions provide a fine information on the conformational properties of polymer chains such as branching and chain flexibility. Osaki and Nemoto made contributions in this field when they joined the group of Prof. Ferry in the University of Wisconsin (17–25, 32).

It was found in the middle of 1950's that the viscosity of polymer concentrates became proportional to a high power, *e.g.* the 3.5–th, of the molecular weight reflecting the formation of chain entanglements when the molecular weight exceeded a critical value M_c. The molecular weight dependence of the steady-state compliance, however, had remained unclear till recently due to various experimental difficulties, and it had long impeded our understanding of the nature of entanglement. To solve this

problem, Osaki and Einaga developed under guidance of Prof. Tamura of this university a new apparatus for creep and creep recovery measurements, and measured the steady-shear compliance of concentrated polystyrene solutions as functions of molecular weight and concentration (26–29, 34, 40). Odani and Nemoto, on the other hand, prepared two series of narrow-distribution polymer samples, poly(*cis*-isoprene) and poly(α-methylstyrene), by anionic polymerization, and carried out precise creep measurements over wide range of molecular weight (35, 55–59). From the results obtained by these two groups, they were able to find that the steady-state compliance became independent of the molecular weight as the molecular weight increased beyond a value M_e, and that the critical molecular weight M_e for the compliance was higher by a factor of two or three than the corresponding M_c for the viscosity.

Our most recent effort in this area was devoted to discriminate a proper type of constitutive equation for nonlinear viscoelasticity from numerous equations so far proposed. We have noticed that the stress relaxation under large deformation provides the most powerful information for the purpose, and have reached after systematic measurements in various types of steady and transient flow at the conclusion that the constitutive equation proposed by Bernstein-Keasley-Zapas (BKZ) is of the most wide applicability (30–31, 37–39, 41–44, 46–54).

The fourth group of the following list includes experimental studies of structure and conformation of various synthetic polymers by means of light scattering, osmotic pressure, viscosity, sedimentation velocity and equilibrium, gel permeation and fluorescence measurements. The polymers studied were poly(methyl ethacrylate) (60), poly(methyl n-butacrylate) (60), poly(1-methoxycarbonyl-1-phenylethylene) (95), poly[oxy(ethylethylene)] (62), poly(α-methylstyrene) (63–65, 72, 76), and poly(styrene) (68, 70, 71, 73, 74). In the first three polymers, it was found that the steric factor in the polymer chain statistics increased monotonously with increasing the size of the substituted groups at the α-carbon of acrylic acid.

The light scattering photometer developed by Utiyama was of quite high precision, by which we were able to reach at 9 degrees of scattering angle without disturbance from stray light (66, 67, 69). This was notable, because the limit of measurements in those days was as high as 20–30 degrees of the scattering angle. Using this apparatus, Utiyama and Tsunashima have shown that in good solvents, the spatial distribution of segments within a molecule really obeys a non-Gaussian statistics due to the excluded volume effect (71), and that poly(α-methylstyrene) consists of large helical sections and displays a thermal transition of highly cooperative nature (72). They carried out light scattering measurements on diblock copolymers of styrene and methyl methacrylate in various solvents including isoreflective solvents for one block, and revealed the effect of solvent nature on the radius of gyration of the visible block and on the size and shape of copolymer aggregates (77, 78). Kurata and coworkers demonstrated that long chain branches in randomly branched polymers can be quantitatively determined only from the intrinsic viscosity and gel permeation data of unfractionated sample (73, 74). Utiyama, Tagata, and Kurata have developed a method for the analysis of sedimentation equilibrium data of polydisperse nonideal systems on the basis of the Fujita theory, which is now accepted as the standard method in ultracentrifugation (68).

Utiyama and Ikehara studied the structure of filamentous bacteriophage fd in various solvents and found that this phage was in a stable intermediate form in 2-chloroethanol (79, 81, 82). Sakato and Utiyama have determined the persistence length of ratskin and earthworm collagens to be 1300 Å (80).

Senior research members worked in this laboratory in the decade are:

Dr. Hisashi Odani (November 1966–)
Dr. Hiroyasu Utiyama (September 1962–)
Dr. Mitsuo Abe (May 1966-April 1967)
Dr. Kunihiro Osaki (April 1967–)
Dr. Nobuo Tagata (July 1967–January 1969)
Dr. Norio Nemoto (April 1968–)
Dr. Kazuyoshi Iwata (April 1969–July 1971)
Dr. Yoshisuke Tsunashima (April 1969–)
Dr. Kuniaki Sakato (April 1970–March 1971)
Dr. Yoshiyuki Einaga (April 1972–March 1973)

Other members consist of two technical assistants, four doctoral thesis students (one from Korea), three master thesis students and two undergraduate students in this academic year (April 1976–March 1977).

Finally Prof. Andrew J. Matheson of University of Essex, England (August-September, 1968), Prof. Stig Claesson of Uppsala University, Sweden (October 1972–January 1973), and Dr. Bengt Finnström of Uppsala University, Sweden (February–March, 1976) were guests of this laboratory for the indicated period, to whom we are grateful for many stimulating discussions on our works.

Publications

(* indicates an article published in Japanese)

I. Polymer Chain Statistics and Hydrodynamics

1. M. Kurata: Ellipsoid Model for Polyelectrolytes, *J. Polymer Sci., Part C,* **No. 15,** 341 (1967).
2. H. Inagaki, H. Suzuki, and M. Kurata: A Semi-Empirical Method for Estimating Unperturbed Dimensions of Chain Polymers from the Intrinsic Viscosity-Molecular Weight Relationship, *ibid., Part C,* **No. 15,** 409 (1967).
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5. K. Iwata: Irreversible Statistical Mechanics of Polymer Chains. I. Fokker-Planck Diffusion Equation, *ibid.,* **54,** 12 (1971).
6. K. Iwata: Irreversible Statistical Mechanics of Polymer Chains. II. Viscosity, *ibid.,* **54,** 1570 (1971).
7. K. Osaki: A Revised Version of the Integrodifferential Equation in the Zimm Theory for Polymer Solution Dynamics, *Macromolecules,* **5,** 141 (1972).
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9. M. Kurata: Thermodynamic Analysis of Mixed Solvent Systems. Part I. Osmotic Pressure and Theta Composition of Solvent, *Bull. Inst. Chem. Res., Kyoto Univ.,* **54,** 112 (1976).

II. Diffusion and Sorption

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11. H. Odani, K. Taira, N. Nemoto, and M. Kurata: Permeation and Diffusion of Gases in Styrene–Butadiene-Styrene Block Copolymers, *Bull. Inst. Chem. Res., Kyoto Univ.*, **53**, 216 (1975).
12. H. Odani, K. Taira, T. Yamaguchi, N. Nemoto, and M. Kurata: Solubilities of Inert Gases in Styrene-Butadiene-Styrene Block Copolymers, *ibid.*, **53**, 409 (1975).
13. H. Odani, K. Taira, N. Nemoto, and M. Kurata: Permeation, Diffusion, and Solution of Gases in SBS Block Copolymer Films, *Denki Kagaku* (*J. Electrochem. and Ind. Phys. Chem.*) **44**, 311 (1976).*

III. Viscoelastic Properties of Polymers

(a) Dilute Solutions

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16. M. Tamura, M. Kurata, K. Osaki, and K. Matsushita: Normal Stress Effect of Dilute Poly(α-methylstyrene) Solutions, *Nihon Kagaku Zasshi*, **89**, 469 (1968).*
17. K. Osaki and J. L. Schrag: Viscoelastic Properties of Polymer Solutions in High-Viscosity Solvents and Limiting High-Frequency Behavior. I. Polystyrene and Poly(α-methylstyrene), *Polymer J.*, **2**, 541 (1971).
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(b) Concentrated Solutions

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(c) **Solids and Melts**

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IV. Structure and Conformation of Macromolecules

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V. Biological Polymers

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VI. Reviews and Books

(a) Reviews

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(b) Books

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